

Exact results for the temperature-field behavior of the thermodynamic Casimir force in a model of film system with a strong surface adsorption

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Abstract. When massless excitations are limited or modified by the presence of material bodies one observes a force acting between them generally called Casimir force. Such excitations are present in any fluid system close to its true bulk critical point. We derive exact analytical results for both the temperature and external ordering field behavior of the thermodynamic Casimir force within the mean-field Ginzburg-Landau Ising type model of a simple fluid or binary liquid mixture. We investigate the case when under a film geometry the boundaries of the system exhibit strong adsorption onto one of the phases (components) of the system. We present analytical and numerical results for the (temperature-field) relief-map of the force in both the critical region of the film close to its finite-size or bulk critical points as well as in the capillary condensation regime below the finite-size critical point.

Keywords: rigorous results in statistical mechanics, classical phase transitions (theory), finite-size scaling

1. Introduction

In a recent article [1], we have derived exact results for both the temperature T and external ordering field h behavior of the order parameter profile and the corresponding response functions – local and total susceptibilities – within the three-dimensional continuum mean-field Ginzburg-Landau Ising type model of a simple fluid or binary liquid mixture for a system with a film geometry $\infty^2 \times L$. In the current article we extend them to derive exact results for the thermodynamic Casimir force within the same model. We concentrate in the region of the parametric space in (T, h) plane close to the critical point of the fluid or close to the demixing point of the binary liquid mixture. We recall that for a simple fluid or for binary liquid mixtures the wall generically prefers one of the fluid phases or one of the components. Because of that in the current article we study the case when the bounding surfaces of the system strongly prefer one of the phases of the system. Since in such systems one observes also the phenomena of the capillary condensation close below the critical point for small negative values of the ordering field $hL = O(1)$, $h < 0$, we will also study the behavior of the force between the confining surfaces of the system in that parametric region. Let us recall that the model we are going to consider is a standard model within which one studies phenomena like critical adsorption [2–15], wetting or drying [12, 13, 16–19], surface phenomena [20, 21], capillary condensation [3, 7, 8, 10, 17, 22, 23], localization-delocalization phase transition [24–26], finite-size behavior of thin films [7, 24, 26–34], the thermodynamic Casimir effect [10, 35–38], etc. Until very recently, i.e. before Ref. [1], the results for the case $h = 0$ were derived analytically [35, 37–39] while the h -dependence was studied numerically either at the bulk critical point of the system $T = T_c$, or along some specific isotherms – see, e.g., [10, 15, 25, 36, 39–41]. In the current article we are going to improve this situation with respect to the Casimir force by deriving exact analytical results for it in the (T, h) plane.

In 1948 [42], after a discussion with Nils Bohr [43], the Dutch physicist H. B. G. Casimir realized that the zero-point fluctuations of the electromagnetic field in vacuum lead to a force of attraction between two perfectly conducting plates and calculated this force. In 1978 Fisher and De Gennes [44] pointed out that a very similar effect exists in fluids with the fluctuating field being the field of its order parameter, in which the interactions in the system are mediated not by photons but by different type of massless excitations such as critical fluctuations or Goldstone bosons (spin waves). Nowadays one usually terms the corresponding Casimir effect the critical or the thermodynamic Casimir effect [34].

Currently the Casimir effect, in its general form, is object of studies in quantum electrodynamics, quantum chromodynamics, cosmology, condensed matter physics, biology and, some elements of it, in nano-technology.

The critical Casimir effect has been already directly observed, utilizing light scattering measurements, in the interaction of a colloid spherical particle with a plate [45] both of which are immersed in a critical binary liquid mixture. Indirectly, as a

balancing force that determines the thickness of a wetting film in the vicinity of its bulk critical point the Casimir force has been also studied in ^4He [46], [47], as well as in ^3He - ^4He mixtures [48]. In [49] and [50] measurements of the Casimir force in thin wetting films of binary liquid mixture are also performed. The studies in the field have also enjoined a considerable theoretical attention. Reviews on the corresponding results can be found in [34, 51, 52].

Before turning exclusively to the behavior of the Casimir force, let us briefly remind some basic facts of the theory of critical phenomena. In the vicinity of the bulk critical point $(T_c, h = 0)$ the bulk correlation length of the order parameter ξ becomes large, and theoretically diverges: $\xi_t^+ \equiv \xi(T \rightarrow T_c^+, h = 0) \simeq \xi_0^+ t^{-\nu}$, $t = (T - T_c)/T_c$, and $\xi_h \equiv \xi(T = T_c, h \rightarrow 0) \simeq \xi_{0,h} |h/(k_B T_c)|^{-\nu/\Delta}$, where ν and Δ are the usual critical exponents and ξ_0^+ and $\xi_{0,h}$ are the corresponding nonuniversal amplitudes of the correlation length along the τ and h axes. If in a finite system ξ becomes comparable to L , the thermodynamic functions describing its behavior depend on the ratio L/ξ and take scaling forms given by the finite-size scaling theory. For such a system the finite-size scaling theory [31–34, 51, 53] predicts:

- For the Casimir force

$$F_{\text{Cas}}(t, h, L) = L^{-d} X_{\text{Cas}}(x_t, x_h); \quad (1)$$

- For the order parameter profile

$$\phi(z, T, h, L) = a_h L^{-\beta/\nu} X_\phi(z/L, x_t, x_h), \quad (2)$$

where $x_t = a_t t L^{1/\nu}$, $x_h = a_h h L^{\Delta/\nu}$. In Eqs. (1) and (2), β is the critical exponent for the order parameter, d is the dimension of the system, a_t and a_h are nonuniversal metric factors that can be fixed, for a given system, by taking them to be, e.g., $a_t = 1/[\xi_0^+]^{1/\nu}$, and $a_h = 1/[\xi_{0,h}]^{\Delta/\nu}$.

2. The Ginzburg-Landau mean-field model and the Casimir force

2.1. Definition of the model

Here, as in [1], we consider a critical system of Ising type in a parallel film geometry described by the minimizers of the standard ϕ^4 Ginzburg-Landau Hamiltonian

$$\mathcal{F}[\phi; \tau, h, L] = \int_0^L \mathcal{L}(\phi, \phi') dz, \quad (3)$$

where

$$\mathcal{L} \equiv \mathcal{L}(\phi, \phi') = \frac{1}{2} \phi'^2 + \frac{1}{2} \tau \phi^2 + \frac{1}{4} g \phi^4 - h \phi. \quad (4)$$

Here L is the film thickness, $\phi(z|\tau, h, L)$ is the order parameter assumed to depend on the perpendicular position $z \in (0, L)$ only, $\tau = (T - T_c)/T_c (\xi_0^+)^{-2}$ is the bare reduced temperature, h is the external ordering field, g is the bare coupling constant and the primes indicate differentiation with respect to the variable z .

2.2. Basic expression for the Casimir force

The thermodynamic Casimir force is the excess pressure over the bulk one acting on the boundaries of the system which is due to the finite size of the system. To derive this excess pressure there are several ways but probably the most straightforward one is to apply the corresponding mathematical results of the variational calculus. For example, following Gelfand and Fomin [54, pp. 54–56] it is easy to show that the functional derivative of \mathcal{F} with respect to the independent variable z at $z = L$ is

$$-\left(\frac{\delta\mathcal{F}}{\delta z}\right)\Big|_{z=L} = -\left(\phi'\frac{\partial\mathcal{L}}{\partial\phi'} - \mathcal{L}\right)\Big|_{z=L}. \quad (5)$$

Having in mind Eq. (4), one derives explicitly

$$\begin{aligned} -\left(\frac{\delta\mathcal{F}}{\delta z}\right)\Big|_{z=L} &= \left(\frac{1}{2}\phi'^2 - \frac{1}{4}g\phi^4 - \frac{1}{2}\tau\phi^2 + h\phi\right)\Big|_{z=L} \\ &\equiv p_L(\tau, h). \end{aligned} \quad (6)$$

This derivative has the meaning of a force acting on the surface of the system at $z = L$ and, since \mathcal{F} is normalized per unit area, it has a meaning of a pressure acting on that surface. That is why, the notation $p_L(\tau, h)$ is used. Another common way to proceed is to use the apparatus based on the stress tensor operator see, e.g., [36], [55] and [56]

$$T_{kl} = \frac{\partial\mathcal{L}}{\partial(\partial_l\Phi)}(\partial_k\Phi) - \delta_{kl}\mathcal{L}. \quad (7)$$

It is elementary to check that the expression in the parentheses in the right-hand-side of Eq. 6 coincides with T_{zz} component of the stress tensor. In [1], we have shown that this expression is a first integral of the considered system, and therefore T_{zz} and $p_L(\tau, h)$ do not depend on the coordinate z at which they are calculated.

In the bulk system, the gradient term in \mathcal{L} is absent and instead of p_L one obtains

$$p_b(\tau, h) = -\frac{1}{4}g\phi_b^4 - \frac{1}{2}\tau\phi_b^2 + h\phi_b, \quad (8)$$

where ϕ_b is the order parameter of the bulk system. Clearly, ϕ_b is determined by the cubic equation $-\phi_b[\tau + g\phi_b^2] + h = 0$, ϕ_b being such that $\mathcal{L}_b = \frac{1}{2}\tau\phi_b^2 + \frac{1}{4}g\phi_b^4 - h\phi_b$ attains its minimum. Now, one can immediately determine the Casimir force as

$$F_{\text{Cas}}(\tau, h, L) = p_L(\tau, h) - p_b(\tau, h). \quad (9)$$

When $F_{\text{Cas}}(\tau, h, L) < 0$ the excess pressure will be inward of the system that corresponds to an *attraction* of the surfaces of the system towards each other and to a *repulsion* if $F_{\text{Cas}}(\tau, h, L) > 0$.

In the light of the above it is evident that once the order parameter profile ϕ is known in analytic form for given values of the parameters τ and h , then the corresponding Casimir force is determined exactly. It is noteworthy that the above expressions do not depend on the specific choice of the boundary conditions. In the current article we specialize to the so-called $(+, +)$ boundary conditions under which one requires that $\lim \phi(z)|_{z \rightarrow 0} = \lim \phi(z)|_{z \rightarrow L} = +\infty$. The exact solution for $\phi(z, \tau, h, L)$ for this case has been determined in [1]. In what follows we will study the properties of the force $F_{\text{Cas}}(\tau, h, L)$ using this exact solution.

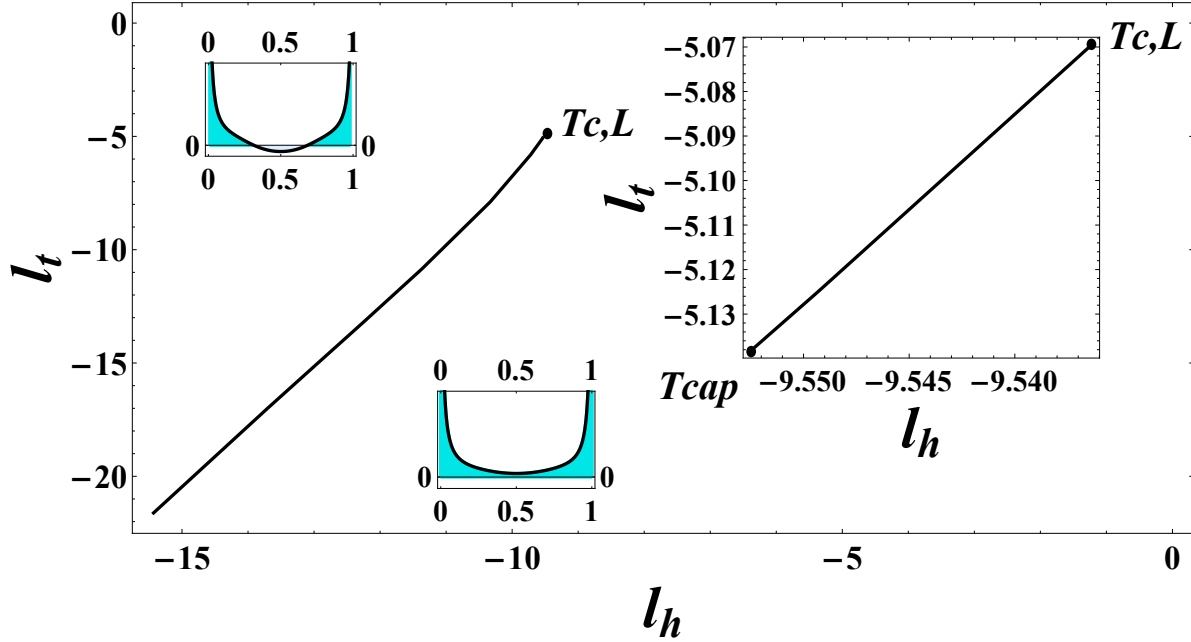


Figure 1: Phase diagram. The critical point of the finite system is $T_{c,L} = (l_t^{(c)}, l_h^{(c)}) = (-5.06935, -9.53633)$. The inset on the right shows the pre-capillary-condensation curve, determined in [1], where above $T_{cap} = (-5.13834, -9.55252)$ and below $T_{c,L}$ the jump the order parameter at the middle of the system is from a less dense gas to a more dense one. For $T \leq T_{cap}$ the system jumps from a "gas" to a "liquid" state.

3. Exact results for the Casimir force

Since the thermodynamic Casimir force is normally presented in terms of the scaling variables

$$l_t \equiv \text{sign}(\tau) L/\xi_t^+ = \text{sign}(\tau) L\sqrt{|\tau|}, \quad (10)$$

$$l_h \equiv \text{sign}(h) L/\xi_h = L\sqrt{3}(\sqrt{g}h)^{1/3}, \quad (11)$$

in the remainder we are going to use such variables as the basic parameters determining the behavior of the force. In the above we have taken into account that for the model considered here $\xi_{0,h}/\xi_0^+ = 1/\sqrt{3}$ [36], $\nu = 1/2$ and $\Delta = 3/2$. The phase diagram of this model has been studied in details in [1]. Here, for the convenience of the reader, it is depicted in figure 1 in terms of the scaling variables l_t and l_h .

3.1. Exact analytical results for the Casimir force

In terms of the scaling variables given in equations (10) and (11), the value $p_L(\tau, h)$ of the first integral, see Eq. (6), becomes

$$p_L(\tau, h) = \frac{1}{gL^4} p(l_t, l_h), \quad (12)$$

where the constant $p(l_t, l_h)$ is

$$p(l_t, l_h) = X'^2 - X^4 - \text{sign}(l_t) l_t^2 X^2 + \frac{2}{3\sqrt{6}} l_h^3 X. \quad (13)$$

Here

$$X(\zeta|l_t, l_h) = \sqrt{\frac{g}{2}} L^{\beta/\nu} \phi(z) \quad (14)$$

is the scaling function of the order parameter ϕ , $\beta = 1/2$ and hereafter the prime means differentiation with respect to the variable $\zeta = z/L$, $\zeta \in [0, 1]$. Similarly, for the bulk system, see Eq. (8), one has

$$p_b(\tau, h) = \frac{1}{gL^4} p_b(l_t, l_h), \quad (15)$$

where

$$p_b(l_t, l_h) = -X_b^4 - \text{sign}(l_t) l_t^2 X_b^2 + \frac{2}{3\sqrt{6}} l_h^3 X_b. \quad (16)$$

From Eqs. (12) and (15) for the Casimir force (9) one obtains

$$F_{\text{Cas}}(\tau, h, L) = \frac{1}{gL^4} X_{\text{Cas}}(l_t, l_h), \quad (17)$$

where its scaling function X_{Cas} is

$$X_{\text{Cas}}(l_t, l_h) = p(l_t, l_h) - p_b(l_t, l_h). \quad (18)$$

Given l_t and l_h , the determination of $p_b(l_t, l_h)$ is evident, while $p(l_t, l_h)$ is given by the expression

$$p(l_t, l_h) = x_m \left(\frac{2}{3\sqrt{6}} l_h^3 - x_m^3 - \text{sign}(l_t) l_t^2 x_m \right), \quad (19)$$

see Eq. (3.15) in [1]. As shown in [1], x_m is to be determined from

$$12\wp\left(\frac{1}{2}; g_2, g_3\right) - \text{sign}(l_t) l_t^2 - 6x_m^2 = 0 \quad (20)$$

so that it gives rise to a continuous order parameter profile in the interval $(0, 1)$, and satisfies the condition

$$6\sqrt{3} x_m (\text{sign}(l_t) l_t^2 + 2x_m^2) - \sqrt{2} l_h^3 > 0. \quad (21)$$

In Eq. (20) $\wp(\xi; g_2, g_3)$ is the Weierstrass elliptic function whose invariants g_2 and g_3 are given by

$$g_2 = \frac{1}{12} l_t^4 + p(l_t, l_h), \quad (22)$$

$$g_3 = -\frac{1}{216} [l_h^6 + l_t^6 - 36 p(l_t, l_h) l_t^2]. \quad (23)$$

Thus, in order to determine $p(l_t, l_h)$ for the regarded $(+, +)$ boundary conditions at given values of the parameters l_t and l_h , one should find all the solutions x_m of the transcendental equation (20) which meet the above requirements. If there is more than

one such solution x_m , as explained in detail in [1], we take that one which leads to an order parameter profile that corresponds to the minimum of the energy functional (3)

$$\mathcal{E} = \frac{1}{gL^4} \int_0^1 f(X, X') d\zeta, \quad (24)$$

where

$$f(X, X') = X'^2 + X^4 + \text{sign}(l_t) l_t^2 X^2 - \frac{2}{3\sqrt{6}} l_h^3 X. \quad (25)$$

The precise mathematical procedure how this can be achieved, despite the divergence of the energy, is also explained in details in [1]. Let us note that x_m has a clear physical meaning - it is the value of the scaling function of the order parameter profile at the middle of the system, i.e., $X(1/2|l_t, l_h) = x_m(l_t, l_h)$.

From Eqs. 16 and 19, once X_b and x_m are determined, the scaling function of the Casimir force takes the form

$$X_{\text{Cas}}(l_t, l_h) = X_b^4 - x_m^4 + \text{sign}(l_t) l_t^2 (X_b^2 - x_m^2) - \frac{2}{3\sqrt{6}} l_h^3 (X_b - x_m). \quad (26)$$

When $h = 0$, i.e. $l_h = 0$, the behavior of the Casimir force under $(+, +)$ boundary conditions has been analytically studied in [35] and [38]. In [38] the value of the so-called Casimir amplitude, i.e., the result for $l_t = l_h = 0$ is obtained, while in [35] the behavior of the force as a function of l_t has been studied.

When $h \neq 0$ the behavior of the Casimir force has been studied only numerically. In Refs. [36] and [56] it has been obtained only for $T = T_c$ for some chosen values of l_h . Below we present its behavior as a function of both l_t and l_h in (l_t, l_h) plane by evaluating numerically the analytical expressions given above.

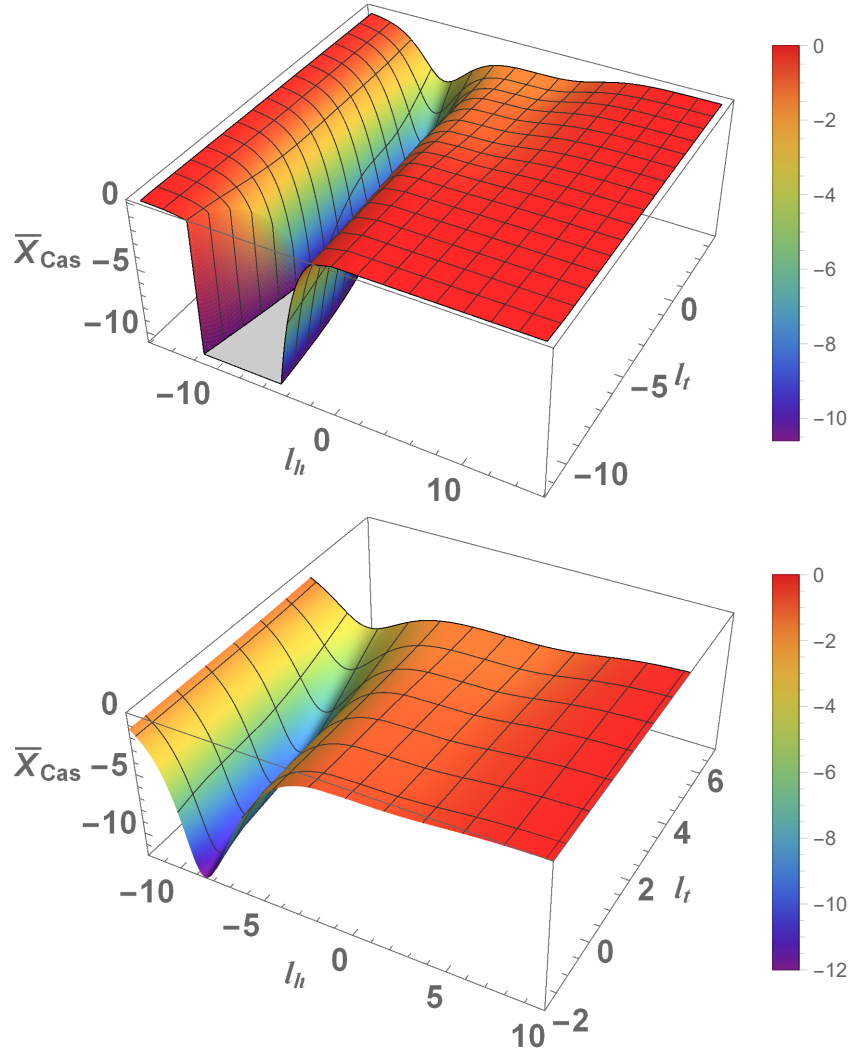
3.2. Numerical evaluation of the analytical expressions

Using the derived exact analytical expressions described above in the current section we determine the Casimir force in the critical and in the capillary condensation regimes. It should be pointed out that the solutions x_m of the transcendental equation (20) that correspond to certain values of the parameters l_t and l_h are to be obtained numerically identifying by inspection those of them that obey the conditions formulated above.

The behavior of the normalized finite-size scaling function $\bar{X}_{\text{Cas}}(l_t, l_h) \equiv X_{\text{Cas}}(l_t, l_h)/|X_{\text{Cas}}(0, 0)|$ of the Casimir force is shown in figures 2 and 3.

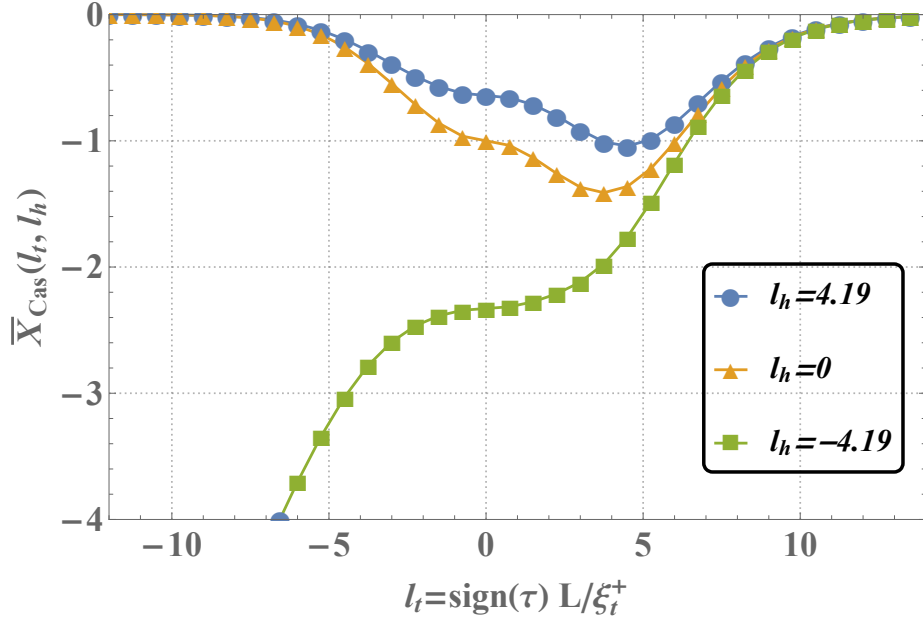
The relief map of the Casimir force, as a function of both l_t and l_h , is shown in figure 2 where the upper part presents the force in a larger scale, while the lower one is a blow up of the region close to the bulk critical point. The only other model we are aware of where such a relief map as a function of both relevant scaling variables is available is that one of the three-dimensional spherical model under periodic boundary conditions [57, 58]. One observes a valley in this map with its deepest part around the phase line of the finite system.

Figures 3a and 3b present cross-sections of the foregoing 3d figures for given fixed values of l_t , or l_h , as a function of l_h , or l_t , respectively. Figure 3a shows the behavior

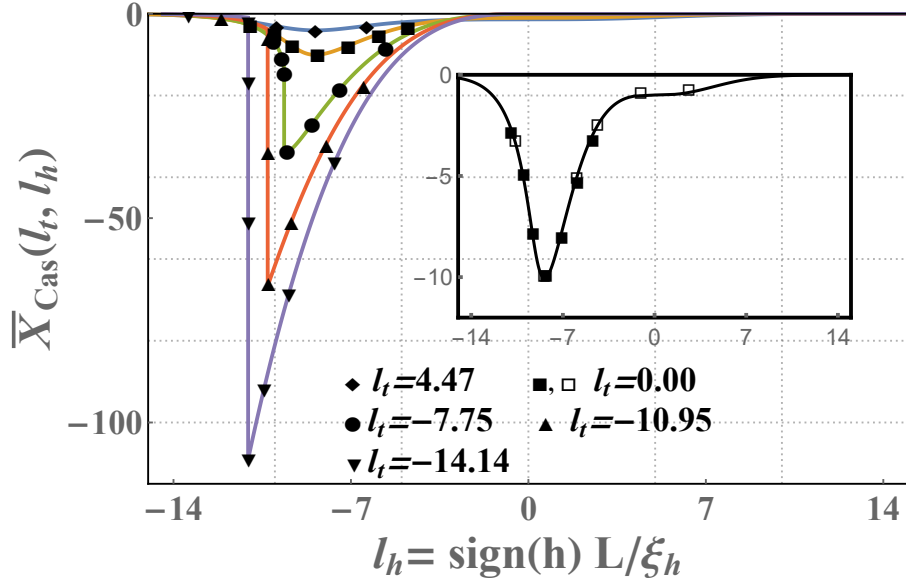
Figure 2: Plots of Casimir force as a function of both l_t and l_h .

of \bar{X}_{Cas} as a function of l_t for $l_h = 0, \pm 4.19$. Note that \bar{X}_{Cas} is negative and for $l_h = 0$ has a minimum at $l_t = 3.749$ above T_c , as in the case of the $2d$ Ising model [59]. The value of the minimum is 1.411 times deeper than the corresponding value of the force at $T = T_c$, which agrees with the results of [35].

Figure 3b depicts the behavior of \bar{X}_{Cas} as a function of l_h for $l_t = 4.47, 0, -7.75, -10.95, -14.14$. Note that the minimum of the function $\bar{X}_{\text{Cas}}(0, l_h)$ is again negative, it is attained at $l_h = -8.405$, and is 10.052 times deeper than the corresponding value at the bulk critical point. The markers on the curves, including the inset curve representing the blow-up in the case $l_t = 0$, show an excellent agreement of the numerical results obtained in [36] (filled markers), and in [56] (empty squares) with the analytic results (solid lines) presented there.



(a) The dependence of the normalized finite-size scaling function $\bar{X}_{\text{Cas}}(l_t, l_h)$ of the Casimir force on the scaling variable l_t for three values of the scaling variable l_h : $l_h = 0$, $l_h = \pm 4.19$.



(b) The dependence of the normalized thermodynamic Casimir force $\bar{X}_{\text{Cas}}(l_t, l_h)$ on the field scaling variable l_h for several values of the temperature scaling variable l_t .

Figure 3: Plots of cross-sections of the Casimir force for given fixed values of l_t , or l_h , as a function of l_h , or l_t , respectively.

4. Discussion and concluding remarks

We have derived exact analytical results for the thermodynamic Casimir force, see Eq. (26), in a widely used model in the theory of phase transitions. In this model, the value x_m of the order parameter in the middle of the system is a solution of Eq. (20) obeying the condition (21). If there is more than one solution x_m satisfying the above requirements, we take the one that leads to an order parameter profile corresponding to the minimum of the energy functional (24), as explained in details in [1]. The precise mathematical procedure how this can be achieved, despite the divergence of the energy, is also explained in [1]. The obtained results allow us to plot the relief map of the force, see figure 2, as a function of the both relevant scaling variables – the temperature and field. Figures 3a and 3b present cross-sections of the behavior of the force for given fixed values of l_t , or l_h , as a function of l_h , or l_t , respectively. The comparison there of the numerical evaluation of our analytical expressions with the available numerical results shows an excellent agreement between each other. Finally, let us recall that the mean-field results also serve as a starting point for renormalization group calculations [21, 35, 53]. Thus, our results shall be helpful for such future analytical studies on the thermodynamic Casimir force. Finally, let us also remind that in physical chemistry and, more precisely, in colloid sciences fluid mediated interactions between two surfaces or large particles are usually referred to as solvation forces or disjoining pressure [3, 59, 60]. Thus, our results can be also considered as pertaining to a particular case of such forces when the fluid is near its critical point.

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